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Correlated charged donors and strong mobility enhancement in a two-dimensional electron gas

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Using *selectively doped* GaAs-$Al_{0.37}Ga_{0.63}As$ and GaAs-AlAs heterostructures, we provide conclusive evidence for correlations among charged donors, DX^--d^+ in $Al_{0.37}Ga_{0.63}As$ and d^+-d^0 in AlAs, which strongly enhance the mobility of a two-dimensional electron gas (2DEG), residing 10 nm away from the charged donors. The determination was accomplished by changing, in the same structure, the extent of correlations without affecting the total number of charged donors and density of the 2DEG. An approximate theory accounts for the measured mobilities.

The motion of *cold* electrons in a perfectly periodic potential, attained in an ideal crystal, is purely ballistic. In doped semiconductors, however, donor atoms are spatially random, thus destroying the periodicity of the crystal and leading to scattering. If donor impurities were spatially correlated, electron mobilities could be extremely high. Since this is difficult to realize, *selectively doped* heterostructures supporting two-dimensional electron gases (2DEG's) are employed. They enable high-mobility transport of electrons in pure GaAs, for example, because the donors are situated in a neighboring $Al_xGa_{1-x}As$ and are spatially separated from the GaAs by an undoped $Al_xGa_{1-x}As$ *spacer*, as seen in Fig. 1(a).[1,2] Still, for spacers on the order of some 30 nm or less, the donor impurities limit the low-temperature mobility. We have chosen this system to look for correlations in the *charged* donors since it is easy to change, *in situ*, the net charge in the donor and 2DEG layers, thus affecting the correlations.

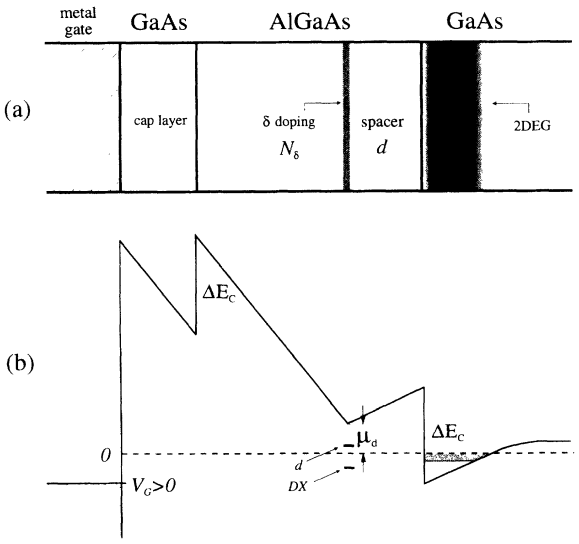

FIG. 1. (a) Layer configuration of the δ-doped heterostructure supporting a 2DEG. (b) Energy band diagram of the heterostructure under $V_G > 0$. The two relevant donor levels are d and DX, and μ_D is the chemical potential associated with the donor system.

The effect of spatial correlations was studied theoretically[3–12] and experimentally[13–17] and various models were proposed. Small enhancement in mobilities were observed in bulk samples,[13–16] however, in 2D systems correlations were only conjectured by comparing a few different structures.[17] It is quite illuminating to look at the expression for the low-temperature momentum relaxation rate of the electrons $1/\tau_t$ (with mobility $\propto \tau_t$), given by the familiar Fermi golden rule,[18]

$$\frac{1}{\tau_t} = \frac{2\pi e^2}{\hbar} \sum_{\mathbf{q}\neq 0} |\langle \mathbf{k}_F + \mathbf{q}|\phi(\mathbf{r})|\mathbf{k}_F\rangle|^2 \delta(E_{\mathbf{k}_F+\mathbf{q}} - E_{\mathbf{k}_F}) \times(1-\cos\theta)\ , \tag{1}$$

where $\langle \mathbf{k}_F + \mathbf{q}|$, and $\langle \mathbf{k}_F|$ are the scattered and initial electronic eigenfunctions with $\mathbf{k}_F$ and $\mathbf{q}$ the Fermi and exchanged wave vectors, $\phi(\mathbf{r})$ is the screened potential produced by the doping layer at the plane of the 2DEG, θ is the scattering angle associated with $\mathbf{q}$, and e and $\hbar$ are the known constants. This known representation can be expressed also in terms of the *correlation function* $\tilde{\phi}(\mathbf{r}) = \int d\mathbf{r}'\phi(\mathbf{r}+\mathbf{r}')\phi(\mathbf{r}')$ of the potential. For the matrix element squared in Eq. (1) we find $|\langle \mathbf{k}_F + \mathbf{q}|\phi(\mathbf{r})|\mathbf{k}_F\rangle|^2 = (1/A^2)\tilde{\phi}(\mathbf{q})$, where $\tilde{\phi}(\mathbf{q})$ is the Fourier transform of the correlation function and A is the area, showing the strong dependence of τ_t on correlations among donors. For example, for a random distribution of impurities $\tilde{\phi}(\mathbf{q}) = N_\delta A|\varphi(\mathbf{q})|^2$, with $N_\delta A$ the total number of ionized donors and $\varphi(\mathbf{q})$ the Fourier transform of the screened potential of a single donor.[18] Experiment indeed shows that the calculated mobilities for uncorrelated potentials can be, in many cases, substantially smaller than the mobility measured experimentally, possibly resulting from spatial charge correlations among the donor impurities.

According to a recently proposed *negative-U* model,[19,20] any isolated group-IV donor in III-V compound semiconductor can be in two different configurations: a shallow, hydrogenic donor (d^+, d^0) at a substitutional site, and a DX-like, deep donor (DX^-, that is, a d^+ donor binding two electrons), displaced along the $\langle 111\rangle$ direction away from the substitutional site.[21]

In Si-doped $Al_xGa_{1-x}As$, the states associated with the DX states are metastable and enter the gap for $x \geq 0.2$. These energy states, related to the number of Al atoms in the bond, follow the L band with a maximal activation energy of some 160 meV at the Γ-X crossover point ($x \approx 0.37$).[22] Their metastability is exhibited mainly by their charge persistence below some *freeze-out* temperature (around 130 K for $x \approx 0.37$), that can be explained in terms of repulsive *configurational* (electronic-elastic) barriers for emission and capture of electrons.

We exploit this metastability to change the net charge and the correlations within the donor layer at elevated temperatures and *freeze* them by cooling to low temperatures. This affects the low-temperature mobility, which serves as a sensor for charge correlations. We find an enhancement in the mobility of the 2DEG by at least a factor of 6, for the same electron density, when correlations among donors are established. Moreover, with these experiments we also verify that the charge of the DX-like Si donors, in their ground state, is negative, agreeing with the negative-U model.

Heterostructures of GaAs-$Al_{0.37}Ga_{0.63}As$ were grown by molecular-beam epitaxy on (100) semi-insulating GaAs substrates (GaAs-AlAs samples will be discussed later). The 2DEG was supplied by a sheet of Si doping (δ doping) (Ref. 23) in the Al-Ga-As with density $N_\delta = 7.5 \times 10^{12}$ cm^{-2} and spacer thickness $d = 10$ nm (Fig. 1). Substrate temperature was low in order to minimize Si out diffusion from the δ doping plane. Metal gated Hall bars were fabricated by a conventional photolithographical process using thermal, rather than electron beam, evaporation of the metal gate, in order to minimize mobility degradation.[24] The mobility, single-particle scattering rate, and density of the 2DEG were measured at 1.4 K via Hall and Shubnikov–de Haas (SdH) measurements, for different ratios of $N(d^+)/N(DX)$, where N stands for the density of the corresponding donors. This was attained by applying a gate voltage, named cooling voltage V_C, at temperatures *above the freeze-out temperature*, and maintaining it while cooling the sample to 1.4 K; thus freezing the donors configuration established at high temperatures (as V_C increases the relative population of DX centers increases). The concentration of the 2DEG, n_s, was then changed at 1.4 K by varying the gate voltage V_G without affecting the frozen concentration of the DX centers. This *thermal cycle* was repeated for different V_C's.

Measuring the electron density, after different thermal cycles, yielded the respective branches shown in Fig. 2. Each branch has a *linear regime* at small V_G's and a *saturation* regime at large V_G's. The linear regime is a manifestation of a constant capacitance per unit area C_G to the plane of the 2DEG, namely, the areal density of the 2DEG is given by $en_s = C_G(V_G - V_D)$, where V_D is the depletion voltage [$n_s(V_D) = 0$, seen in Fig. 2]. The saturation regime, however, indicates that electrons *flood* the shallow d^+ donor sites and screen the field of the gate.

The measured mobilities are plotted as a function of n_s in Fig. 3, for the various thermal cycles. Note the mobility drop at $n_s = 6 \times 10^{11}$ cm^{-2} that is known to be related to an onset of a second subband transport.[25]

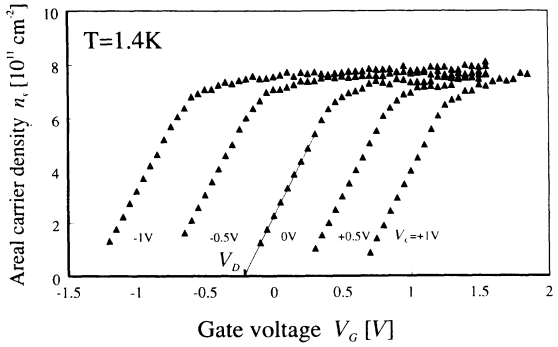

FIG. 2. Areal carrier density of the 2DEG vs gate voltage for several *thermal cycles* with different values of cooling gate voltages V_C. In the linear regime the charge in the donor layer is constant, and in the saturation regime the gate is screened. The depletion voltage, namely, $n_s(V_D) = 0$, is shown for a particular thermal cycle.

The charge density within the donor layer Q_δ is *constant* for each thermal cycle, in the linear regime, and depends strongly on V_C. For the first three thermal cycles it is calculated, using the appropriate V_D's and structure parameters, $Q_\delta/e = 5.9 \times 10^{11}$, 1.9×10^{12}, and 3.5×10^{12} cm^{-2} for $V_C = 1$, 0, and -1 V, respectively. Even though Q_δ varies by almost an *order of magnitude* the mobility is found to be *almost independent* on these three thermal cycles. This observation provides an important insight into the charge state of the DX donors. In the model where the donors are in DX^0-d^+ states, only the positively charged donors d^+ contribute to scattering. This model predicts a substantial decrease in mobility with decreasing V_C due to an increase in $N(d^+)$, contrary to the observed results. In the DX^--d^+ model,[19,20] however, the number of scatterers is constant and equal to $N(d^+) + N(DX^-) = N_\delta$, and thus, the mobility is expected to be approximately the *same* for all V_C's, as indeed observed.

If indeed charge correlations were to set in, when would

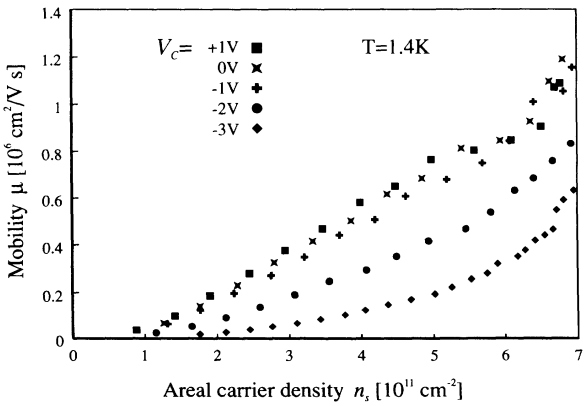

FIG. 3. Mobility vs areal carrier density of the 2DEG for the different *thermal cycles*. The mobility drops for $V_C < -1$ V. Note the onset of second subband transport for $n_s > 6 \times 10^{11}$ cm^{-2}.

we expect them to be significant? It is convenient to define by η the fraction of the donors which is negative, $\eta = N(DX^-)/N_\delta$, with $0 \leq \eta \leq 1$. The value $\eta = 0$ ($\eta = 1$) is realized by thermal cycles with very negative (positive) V_C's, making all donors d^+-like (DX^--like). For these extremes correlations among charges cannot take place because of the random positions of the donors.[26] To calculate η we use the net charge in the doping layer $Q_\delta = e[N(d^+) - N(DX^-)]$, which depends on V_C, and the total doping level $N_\delta = 7.5 \times 10^{12}$ cm^{-2}, and find for $V_C = -3, -2, -1, 0, +1$ V, $\eta = 0.03, 0.15, 0.27, 0.37, 0.46$, respectively. Note, that it is not feasible to reach experimentally $\eta > 0.5$ due to an onset of gate leakage current. So, it is reasonable to believe that correlations might be important around $\eta \approx 0.5$ ($V_C = 0, +1$ V) and diminish for $\eta \leq 0.2$ ($V_C \leq -2$ V), as indeed observed in the large mobility drop seen in Fig. 3.

Correlations can be also partly destroyed via photoexcitation of electrons out of the DX^- centers.[13] Being a spatially random process, photoexcitation is expected to create random, persistent d^+ or d^0 centers at low temperatures, and thus reduce correlations (even though η can still be far from its extremes). To explore this effect the thermal cycles with $V_C = +0.5$ and $+1$ V, having large η and low $n_s(V_G = 0$ V), were followed by controlled illumination with a sequence of short optical pulses, applied by an infrared light emitting diode, at $V_G = 0$ V and 1.4 K. The process continued until n_s sufficiently increased to the same value as in the thermal cycle with $V_C = 0$ V. Even though $n_s(V_G)$, Q_δ, N_δ, and η are *identical* to these after the *dark* thermal cycle with $V_C = 0$ V, the mobilities are lower by up to a factor of 6 (see Fig. 4). The more dramatic effect observed when illumination followed the thermal cycle with $V_C = +1$ V is a direct result of the longer illumination needed to restore $n_s(V_G = 0$ V), and thus the higher randomization of charges.

We discuss now the theoretical implications of correlations among charged donors, based on the DX^- model we have substantiated above. We derive the Fourier transform of the correlation function $\tilde{\phi}(\mathbf{q})$ in terms of the average density of positive and negative scatterers and the pair correlation functions $g_{ab}(r)$. These are the distribution functions of the conditional probability to have a b-type ion at a distance r from the origin given that an a-type ion is at the origin ($a, b = \pm e$). To calculate $g_{ab}(r)$, the induced charge density of positive and negative ions $\rho(r)$ around a test charge is described in terms of spatially continuous functions, obtained by employing the Boltzmann statistics, $\rho(r) \propto \exp[-e\psi(r)/k_BT]$, in conjunction with the Poisson equation [where k_B is the Boltzmann constant and $\psi(r)$ is the potential]. The continuous charge model may be valid only for minority charges surrounded by majority ones with the opposite sign (η is near its extremes); however, at these extremes, correlations of minority charges are expected to be weak (as discussed above) and scattering is dominated by the randomly distributed, *uncorrelated*, majority charges. On the other hand, near $\eta \approx 0.5$, when correlations of both charges are strong and dominate the mobility, we could not find a theoretical justification for the continuous model, hence leaving its verification to the experiment. Assuming weak correlations, which imply thermal energy at the freeze-out temperature T_f on the order of the Coulomb energy, $e^2\sqrt{N_\delta}/\varepsilon \approx k_BT_f$ (leading to $T_f > 100$ K for our doping level), we find[27]

$$\tilde{\phi}(\mathbf{q}) = N_\delta A \frac{q}{q + \alpha[1 - \exp(-2dq)]} |\varphi(q)|^2 , \qquad (2)$$

where $\alpha = e^2N_\delta/2\varepsilon k_BT_f$. The term $\exp(-2dq)$ arises from the image charges of the donors due to the 2DEG conductive plane. Equation (2) predicts that T_f is *crucial* in determining the extent of correlations (similar to Ref. 3 which treated a DX^0-like model).

Using Eq. (2) in conjunction with the Fang-Howard wave function for the 2DEG (Ref. 1) we obtained the momentum relaxation rate and mobility versus 2DEG density. In Fig. 4 we plot the calculated mobilities for an uncorrelated scattering potential and for a correlated one with $T_f = 130$ K, and compare them with our experimental results. We find an excellent agreement between the calculated *correlated* mobility and the experimental one (for $V_C = 0$ and single band transport). The calculated *uncorrelated* mobility has the lowest value in Fig. 4, suggesting that residual correlations persist even after the thermal cycle with $V_C = -3$ V ($\eta = 0.03$), or after illumination. Note here that our measurements of the single-particle relaxation times (not shown here), deduced from the amplitude of the SdH magnetoresistance oscillations,[28] disagreed with our theory even on a qualitative basis.

To further substantiate our results we have fabricated and measured two other types of heterostructures. (a) One has the same AlAs mole fraction but with lower donor concentrations. In general, comparison between different structures is unreliable since the mobility depends strongly on the spacer width, which is not reproducible to the desired accuracy. Hence we compared the relative mobility reduction as correlations were destroyed in each structure. We found for structures with $N_\delta = 7.5 \times 10^{12}$, 2.5×10^{12}, 1.0×10^{12}, and 6.0×10^{11} cm^{-2}, mobility reduction of up to a factor of 6, 1.8, 1.4, and 1.3, respectively. This is expected since Coulom-

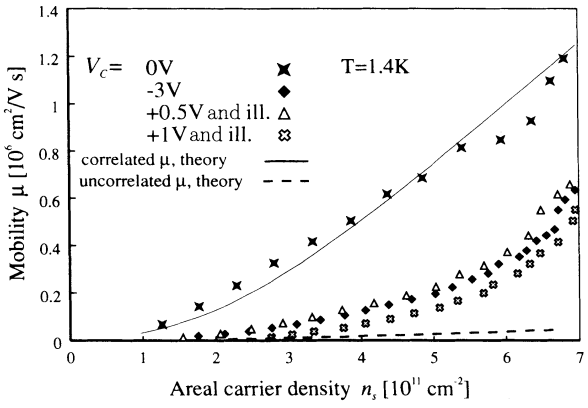

FIG. 4. Comparison of calculated and measured mobilities. The experimental results all lie between the calculated *correlated* and *uncorrelated* mobilities.

bic interactions are weaker, and so are the correlations, when doping is lower. (b) The second heterostructure has pure AlAs supply layers (spacer width also 10 nm). They are expected to have a lower *effective* freeze-out temperature due to the degeneracy of the single DX level with the conduction band leading to a population of nonmetastable, shallow-type donors (d^+, d^0) alone, and thus higher mobilities. Indeed freeze-out effects were not observed and our thermal-cycle-type experiments were not possible. However, the measured mobilities were about *twice* as high in these samples than in the $Al_xGa_{1-x}As$ heterostructures for the same Q_δ (at $T = 1.4$ K, $\mu = 5 \times 10^5$ cm^2/V s for $n_s = 2.5 \times 10^{11}$ cm^{-2}), suggesting a higher degree of correlations.

In summary, by demonstrating a strong mobility enhancement, we give conclusive evidence for existence of correlations among positively and negatively charged donors, coexisting in selectively doped GaAs-Al(Ga)As heterostructures. Moreover, we also prove that the donors can be either metastable DX^--like or shallow, hydrogenic donors, agreeing with the recently predicted negative-U model. The experimental results were compared with a simple theory that seems to account for the enhancement in the mobilities due to correlations.

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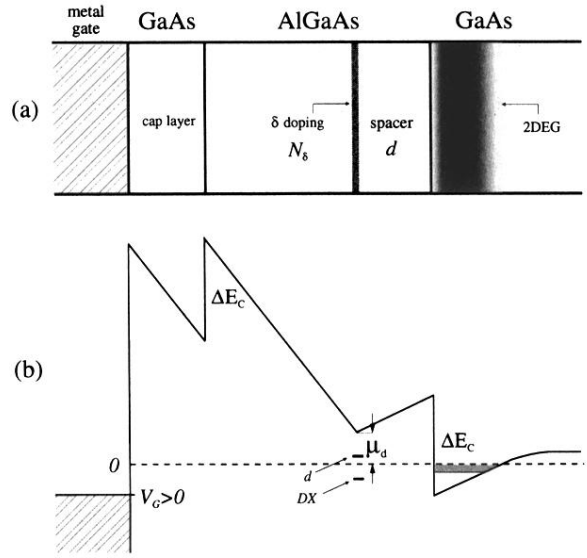
[1] T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).
[2] E. E. Mendez, IEEE J. Quantum. Electron. **QE-22**, 1720 (1986).
[3] A. L. Efros, F. G. Pikus, and G. G. Samsonidze, Phys. Rev. B **41**, 8295 (1990).
[4] R. Lassing, Solid State Commun. **65**, 765 (1988).
[5] J. Van Hall, Superlatt. Microstruct. **6**, 213 (1989).
[6] P. J. Van Hall, T. Klaver, and J. H. Wolter, Semicond. Sci. Technol. **3**, 120 (1988).
[7] B. K. Ridley, Semicond. Sci. Technol. **3**, 111 (1987).
[8] P. Sobkowicz, Z. Wilamowski, and J. Kossut, Semicond. Sci. Technol. **7**, 1155 (1992).
[9] J. Mycielski, Solid State Commun. **60**, 165 (1986).
[10] Don Monroe, Appl. Phys. Lett. **59**, 2293 (1991).
[11] E. P. O'Reilly, Appl. Phys. Lett. **55**, 1409 (1989).
[12] P. J. Van Der Wel, M. J. Anders, L. J. Giling, and J. Kossut, Semicond. Sci. Technol. **8**, 211 (1993).
[13] P. L. Coz, C. Ghezzi, and A. Parisini, Semicond. Sci. Technol. **8**, 13 (1993).
[14] D. K. Maude, Appl. Phys. Lett. **60**, 1993 (1992).
[15] Z. Wilamowski, J. Kossut, T. Suski, P. Wisniewski, and L. Dmowski, Semicond. Sci. Technol. **6**, B34 (1991).
[16] L. H. Dmowski, B. Goutiers, D. K. Maude, L. Eaves, J. C. Portal, and J. J. Harris, Jpn. J. Appl. Phys. **32**, 221 (1993).
[17] P. T. Coleridge, Phys. Rev. B **44**, 3793 (1991).
[18] T. Ando, J. Phys. Soc. Jpn. **51**, 3900 (1982).
[19] D. J. Chadi and K. J. Chang, Phys. Rev. Lett. **61**, 873 (1988).
[20] D. J. Chadi and K. J. Chang, Phys. Rev. B **39**, 10 063 (1989).
[21] J. Makinen, T. Laine, K. Saarinen, P. Hautojarvi, C. Corbel, V. M. Airaksinen, and P. Gilbart, Phys. Rev. Lett. **71**, 3154 (1993).
[22] P. M. Mooney, J. Appl. Phys. **67**, R1 (1990).
[23] E. F. Schubert, J. Vac. Sci. Technol. A **8**, 2980 (1990).
[24] M. J. Rooks, C. C. Eugster, J. A. del Alamo, G. L. Snider, and E. L. Hu, J. Vac. Sci. Technol. B **9**, 2858 (1991).
[25] H. L. Stormer, A. C. Gossard, and W. Wiegmann, Solid State Commun. **41**, 707 (1982).
[26] The distribution of Si sites is not totally random (for example, a minimum distance between two Si atoms exists); however, these correlations are small compared to Coulomb originated correlations.
[27] E. Buks (unpublished).
[28] B. Laikhtman and E. L. Altshuler, Ann. Phys. (to be published).



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